Time-of-Flight Photon Spectroscopy: A Simple Scheme To Monitor Simultaneously Spectral and Temporal Fluctuations of Emission on Single Nanoparticles

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Résumé en anglais	Here we report on a novel scheme for spectral analysis that exploits the wavelength dependence of the time-of-flight of a photon in a dispersive medium. This versatile and cost-effective method, named time-of-flight photon spectroscopy (TOFPS), has the major advantage of being compatible with time-correlated single-photon counting experiments. Consequently, each photon acquired during an experiment is characterized by two parameters, its absolute time of arrival and its color, respectively. As a result, the spectral and temporal fluctuations of the emission of a single nano-object can be derived from a single measurement. As a proof of the concept, we demonstrate in the paper that the method can be used to perform Raman spectroscopy as well as fluorescence spectroscopy. We emphasize that TOFPS proves to be very efficient for improving signal-to-noise ratio in fluorescence correlation spectroscopy measurements by subsequent spectral filtering and to record luminescence spectra from single metallic particles. We demonstrate that the opportunity of simultaneously recording spectral and temporal fluctuations could be used to sort particles of different shapes inside a sample. TOFPS furthermore allows developing a new type of time interval distribution analysis which correlates the time interval between two photons and their corresponding color shift. It is applied to the analysis of the two-photon excited luminescence of a single gold nanorod. This method has a potential for a broad range of applications, among which time-resolved SERS spectroscopy and analysis of the dynamics of emission processes can be handled with new statistical approaches based on the correlation of spectral and temporal fluctuations.
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